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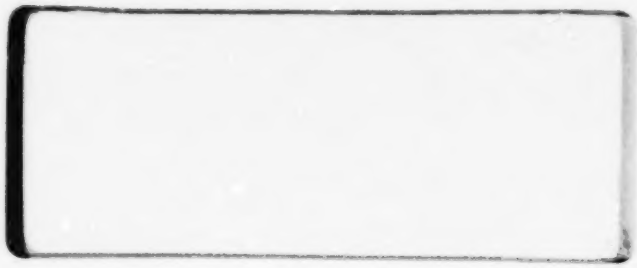

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Report for the

CANADIAN ELECTRICAL ASSOCIATION
Research & Development
Suite 580, One Westmount Square
Montreal, Québec H3Z 2P9

and for the

TRANSPORTATION DEVELOPMENT CENTRE
Complexe Guy Favreau
Suite 601, West Tower
Montréal, Québec H2Z 1X4

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DEVELOPMENT AND TESTING OF A LARGE LITHIUM MOLYBDENUM DISULFIDE BATTERY

Prepared by

MOLI ENERGY LIMITED
3958 Myrtle Street
Vancouver, B.C. V5C 4G1

Principal Investigator: Dr. K. Brandt

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17. Abstract

The objective of this project was to demonstrate the feasibility of the scale-up of the lithium molybdenum disulphide ambient temperature rechargeable battery technology to a 0.5 kW-h storage battery. Using the technology developed for the 'C' size cells, 50 A-h cells were constructed and tested. With a battery consisting of 6 cells in series, an energy storage capacity of 686 W-h, a gravimetric energy density of 94 W-h/kg, and a peak power density of 128 W/kg were demonstrated. It was concludes that this technology, with some further improvement, is suitable for traction applications such as electric vehicles.

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17. Résumé

L'objectif de la présente recherche était de montrer la faisabilité d'agrandir la taille de la batterie rechargeable lithium-disulfure de molybdène à température ambiante pour lui faire atteindre la puissance de 0,5 kWh. Tirant parti de la technique étudiée pour les batteries de taille C, des cellules de 50 Ah chacune ont été construites et essayées. Des batteries formées de six cellules en série ont été réalisées, d'une capacité de 686 Wh et donnant une énergie massique de 94 Wh/kg et une puissance massique de pointe de 128 W/kg. Une fois mises au point, ces batteries pourraient servir à la traction routière électrique.

18. Mots-clés	19. Diffusion
Batteries, rechargeable, lithium, agrandissement, énergie massique, véhicules électriques.	Le Centre de développement des transports dispose d'un nombre limité d'examplaires

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SUMMARY

Moli Energy Limited has developed a rechargeable battery technology based on the electrochemical couple lithium and molybdenum disulfide in a non-aqueous electrolyte. 'AA' and 'C' size cells are currently being introduced into the market. Their main advantage over other commercially available cells using aqueous electrolytes,e.g. lead acid and nickel cadmium, is the high volumetric and gravimetric energy density. It is this advantage that makes the system very attractive for large scale applications like electric vehicles.

The objective of this contract, which was jointly funded by the Canadian Electrical Association, the Transportation Development Centre, and Moli Energy Limited, was to demonstrate the feasibility of a scaleup of the MOLICEL™ technology to a 'BC' size cell with a capacity of about 50 Ah, and to evaluate the performance of a .5 kWh storage battery consisting of 6 'BC' size cells in series.

The design of the 'BC' cell was based on concepts developed for 'C' size cells. Both cell types are of a jelly roll type construction and the scale-up was achieved by increasing the electrode area by a factor of 22. Due to the similarity of the design, modified 'C' cell assembly equipment could be used, in most cases, for the fabrication of 'BC' cells. When this approach was not feasible, special equipment was designed and built. Test equipment for performance and safety testing of 'BC' cells and batteries had to be either purchased or built.

A total of 18 cells were built and tested. Of these cells, the first 5 were of a first design (type 1), the remaining cells were of an improved design (type 2). The design changes included improved safety vents, and a new electrolyte. In testing, type 2 cells achieved a capacity between 41.5 Ah and 66.5 Ah on the first cycle, depending on the cycling conditions. The average cell voltage on discharge was

1.8 V. The cells were shown to be capable of delivering a continuous current output in excess of 50 A at temperatures of -10°C and above.

Some limited safety testing was performed on single cells. Under some conditions, venting of the cells associated with flame occurred. On short circuit, cells did not vent until 20 minutes into the test. The cells tolerated at least 45% overcharge and 250% of forced discharge without venting.

A battery with 2 cells in series and a battery with 6 cells in series were built and tested. The 6 cell battery had an average voltage of 10.8 V. Depending on the cycling conditions, a capacity of up to 63.5 Ah was achieved. This corresponds to an energy storage capacity of 686 Wh, and a gravimetric energy density of 94 Wh/kg. This represents about 2.5 times the energy density of an advanced lead acid battery. The maximum peak power output of this battery was determined as 932 W, corresponding to a power density of 128 W/kg. Further increases in the energy density are possible by optimizing the cell design. Improvements to the power density, which is somewhat lower than the power density of advanced lead acid, will be possible by optimizing the design of current carrying cell parts.

SOMMAIRE

Moli Energy Limited a mis au point un accumulateur au lithium/disulfure de molybdène, à électrolyte non aqueux, qu'elle a entrepris de commercialiser en versions "AA" et "C". Le principal avantage de cet accumulateur par rapport aux batteries classiques à électolyte aqueux- batteries au plomb et nickel-cadmium - est sa grande capacité en énergie par unité de masse ou de volume, qualité qui est justement recherchée dans les applications demandant une puissance et une autonomie élevées: par exemple, les véhicules électriques.

L'objectif de cette recherche, co-financé par l'Association canadienne de l'électricité, le Centre de développement des transports et Moli Energy Limited, a été de montrer la faisabilité d'adapter la technologie Molicel TM à la fabrication d'éléments de type BC offrant une capacité d'environ 50Ah et d'évaluer un accumulateur 0,5kWh composé de 6 éléments BC montés en série.

La conception de l'élément BC est fondé sur les mêmes principes que les éléments de type C. Les deux sont de type cylindrique à électrode enroulée en spirale et baignant dans un électrolyte gélifié, le gain en énergie et puissance ayant été obtenu en augmentant la surface des électrodes par un facteur de 22. Les caractéristiques constructives des deux éléments étant similaires, on a pu utiliser la même machine, modifiée, pour la fabrication de la plupart des prototypes, mais il a également fallu concevoir et construire une machine spéciale pour les autres. Les appareils d'essais utilisés pour mesurer les performances et contrôler la sécurité des éléments BC ont été achetés ou fabriqués de toutes pièces.

Au total, 18 éléments ont été fabriqués et testés, 5 d'un premier modèle (désigné type 1), et les autres d'un modèle amélioré (type 2). Les améliorations ont porté entre autres sur les évents et l'électrolyte. La capacité des éléments de type 2 a varié entre 41,5 Ah et 66,5 Ah lors du premier cycle, selon la tension de charge-décharge adoptée. La tension moyenne aux bornes des éléments en cours de décharge s'est établie à 1,8 volt. Il a été montré que ces éléments pouvaient débiter un courant de plus de 50 ampères, à -10°C et plus.

Certains éléments ont été soumis à des épreuves limitées de sécurité. Dans certaines conditions, on a noté une montée en pression suffisante pour provoquer l'ouverture de l'évent de sécurité et l'évacuation de gaz enflammés. A l'essai de court-circuit, les évents ne se sont ouverts qu'au bout de 20 minutes. Les éléments ont toléré une surcharge d'au moins 45% et une décharge forcée de 2,5 fois leur capacité nominale, avant ouverture de l'évent.

Un accumulateur à deux éléments en série et un autre à six ont été assemblés et testés. La tension moyenne aux bornes du deuxième a été de 10,8 volt. Suivant la tension de charge-décharge adoptée, une capacité maximum de 63,5 Ah a pu être réalisée. Cela correspond à une capacité en énergie de 686 Wh et une énergie massique de 94 Wh/kg, soit environ 2,5 fois celle d'un accumulateur au plomb perfectionné. Quant à la puissance maximum de crête pouvant être obtenue de cet accumulateur, elle s'établit à 932 W pour une puissance massique de 128 W/kg. L'énergie massique de ce type d'accumulateur peut cependant être améliorée par l'optimisation des éléments tandis que, côté puissance massique, l'amélioration nécessaire pour combler l'écart qui sépare cet accumulateur des modèles au plomb perfectionnés passe par l'optimisation des conducteurs de l'élément.

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1 INTRODUCTION

Moli Energy Limited ('Moli') has, during the past seven years, developed a technology base that is leading to the commercialization of a new family of electrochemical batteries. These batteries utilize the phenomenon of intercalation in a lithium molybdenum disulfide system. The first products developed are a 'C' size cell and an 'AA' size cell, common formats used for many modest scale applications. Large scale production of these cell sizes will begin in spring of 1986.

Test cells of 'C' and 'AA' size produced by Moli Energy and evaluated by Moli and independent laboratories show a number of unique advantages over conventional rechargeable systems like Nickel Cadmium. Moli 'C' cells have about twice the gravimetric energy density of Nickel Cadmium cells, they have a very good charge retention capability of over eight years compared to about 4 months for Nickel Cadmium, and they have a state of charge indicator. Furthermore, they have a high rate capability, perform over a wide ambient temperature range, and are inherently safe at cell temperatures below 100°C.

The nature of the MOLICEL™ technology is such that cells and batteries can be scaled up readily to satisfy the requirement of larger scale applications such as electric vehicles. For this project, a cell size with a capacity of about 50 ampere hours was chosen, because the scale up to this size could be performed with presently available components. The physical size of this cell is close to that of the 'BC' size cell which is a battery industry standard. The largest battery size that was studied was composed of 6 'BC' size cells in series. This size is sufficient to power a small electric vehicle like a wheelchair.

This report describes the 'BC' cell project which was supported by Moli, the Transportion Development Centre, and the Canadian Electrical Association. The report is divided into three main sections. The

first section briefly describes the two designs of the 'BC' cell which were developed by Moli. The cell design was not part of this project. The second section describes the fabrication and test equipment that was developed and built for this project. This section was mainly supported by the Transportion Development Centre. The third section deals with the fabrication of 'BC' size cells and batteries and the results of performance and safety testing. The last part was largely supported by the Canadian Electrical Association.

2 DESIGNS OF THE MOLICEL™ 'BC' SIZE CELL

The MOLICEL™ technology utilizes a non-porous lithium metal anode and a porous molybdenum disulfide cathode. The basic chemistry of this system is the intercalation of lithium into molybdenum disulfide according to the following equation:

$$x \cdot Li + MoS_2 \underset{\text{charge}}{\overset{\text{discharge}}{\langle\text{----------}\rangle}} Li_xMoS_2$$

During charge and discharge of the cells the lithium concentration in the intercalation compound, x, varies between values larger than 0 and smaller than 1.

Cells are built with an excess of lithium metal which also serves as an anode current collector. The current collector for the cathode is a thin metal foil onto which the cathode active material has been deposited forming a highly porous layer with a large surface area. The active material covers only one side of the foil. A microporous polypropylene sheet serves as a separator. The electrolytes that can be used are based on aprotic organic solvents and lithium salts which are soluble in these solvents. An example of a fairly common electrolyte is a 1 molar solution of lithium hexafluoroarsenate ($LiAsF_6$) in propylene carbonate (P.C.).

Cells are built by sandwiching the lithium foil between two cathodes with the active material facing the anode. Two separators separate the active materials. The thickness of this web is about 460 µm. The nominal capacity per unit area of web is approximately 5.7 mA hours/cm^2. For a 'C' size cell, the web is 4.19 cm wide and approximately 90 cm long. The active area, which is twice the web area, is about 754 cm^2. The web is wound spirally onto a mandrel resulting in a jelly roll type structure with a diameter of 2.44 cm. The scaleup to the 'BC' size is performed by increasing the electrode

area and not by increasing the electrode thickness. The 'BC' cell has a jelly roll with a height of 12.4 cm and a diameter of 6.5 cm. The total active area is about 16,700 cm^2. This gives a nominal capacity of 47.6 Ah.

Electrical connections to the current collectors in the jelly roll are made via nickel tabs which are spot welded to the lithium foil and the cathode current collector, respectively. In order to reduce the voltage drop along the current collector, 6 tabs are used for each electrode positioned at constant intervals along the web.

The cell can of the first 'BC' cell size ('BC I'), was made of .48 mm thick stainless steel. The cell cap incorporated a safety vent, a negative terminal, a positive terminal which was insulated from the can by using a glass-to-metal seal, and a fill tube for filling the cell with electrolyte. Anode and cathode tabs were connected to the respective terminals using a screw type connection. The cap was welded into the can using inert gas welding. The cell was filled with an electrolyte consisting of a 1 molar solution of $LiAsF_6$ in P.C.

For the second design, which is shown in Figure 1, improvements were made which were both the result of tests of cells of the type 'BC I' and of developments of the MOLICEL™ 'C' size cell. Cells of the type 'BC II' had nickel plated mild-steel cans, and a safety vent both in the bottom of the cell and in the top. These vents open a section of can bottom and top at a given pressure by tearing the can material along a 'C' shaped score. The terminals were redesigned in order to reduce cell impedance. An improved electrolyte was used which was developed by Moli using smaller cell sizes in order to improve cell capacity and cell cycle life. The introduction of this electrolyte in 'C' cells doubled the cycle life and increased the usable cell capacity substantially.

Table 1 summarizes all the design features described above.

Table 1
Summary of Features of the Two 'BC' Size Cell Designs

		'BC I'	'BC II'
Cell height	[cm]	15.2	15.2
Cell diameter	[cm]	6.67	6.67
Cell volume	[cm^3]	531	531
Cell weight	[g]	1,300*	1,100
Nominal cell capacity	[A hours]	47.6	47.6
Average cell voltage	[V]	1.85	1.85
Active area	[cm^2]	16,700	16,700
Case material		Stainless Steel	Ni-plated Steel
Safety features		Single vent	Double vent
Electrolyte		Type I	Type II

* including fill tube valve

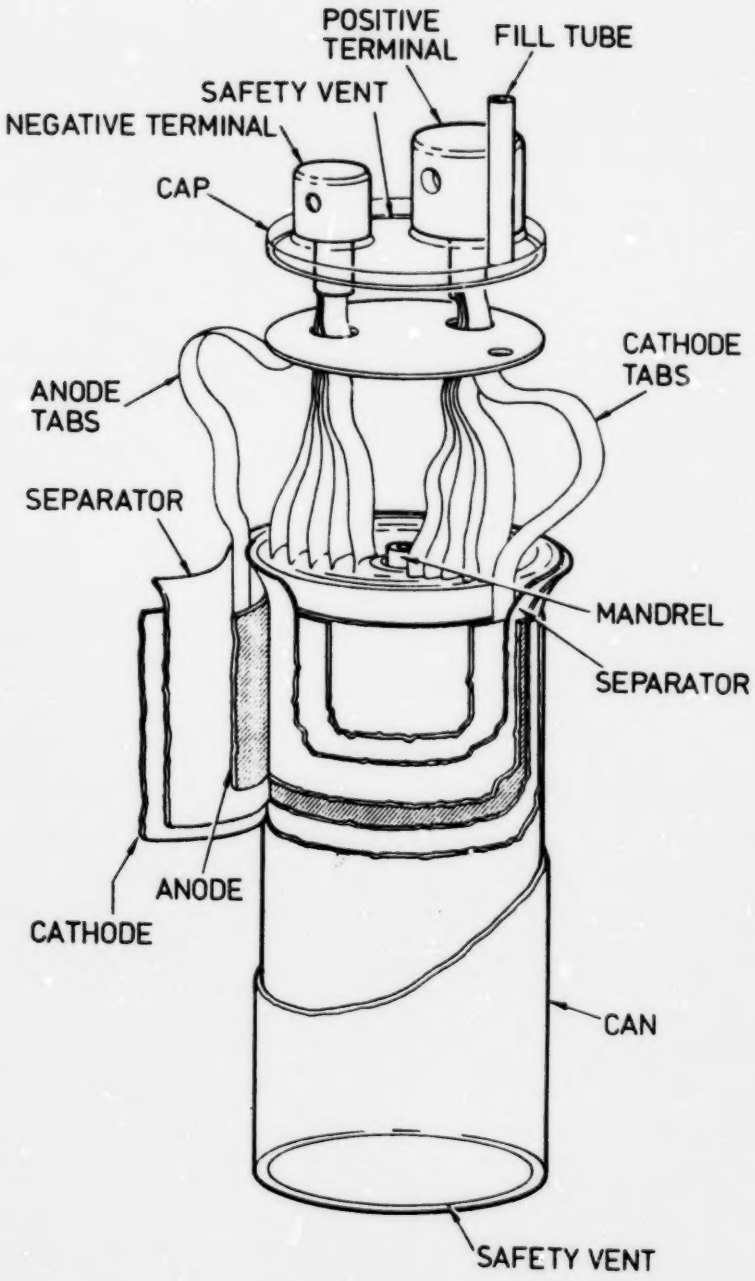

Figure 1: Exploded View of a 'BC' cell, design 'BC II'

3 FABRICATION AND TEST EQUIPMENT

3.1 INTRODUCTION

This section describes the fabrication and test equipment that was needed for the project. Whenever possible, existing equipment, originally designed for the smaller cells was used or adapted for the 'BC' size cells.

Figure 2 shows a block diagram of the cell fabrication. Cathodes were fabricated from raw MoS_2 as received from the molybdenite mines. Purification and other preparation steps for the active material were performed using existing equipment. For fabrication of the 12.4 cm wide cathodes, modifications to the existing cathode machine were necessary. Lithium foil was used as received but had to be respooled onto reels which fit the 'BC' cell winder, which wound the jelly roll from its components.

In the cell assembly step, the jelly roll was inserted into the can, connections from the electrodes to the terminals were made by spot-welding, and the cell cap was then inserted into the can. The cap and can are welded together at the rim using tungsten inert-gas arc welding (T.I.G.).

The cells were assembled dry, that is, without electrolyte, and later were filled with electrolyte through the fill tube which was then sealed off. The cell finishing step comprised an electrochemical treatment of the cathode active material, which resulted in a structurally modified MoS_2 with a greater capacity for lithium intercalation than naturally occuring MoS_2(1).

The following gives a more detailed description of the equipment supplied for 'BC' cell fabrication and testing.

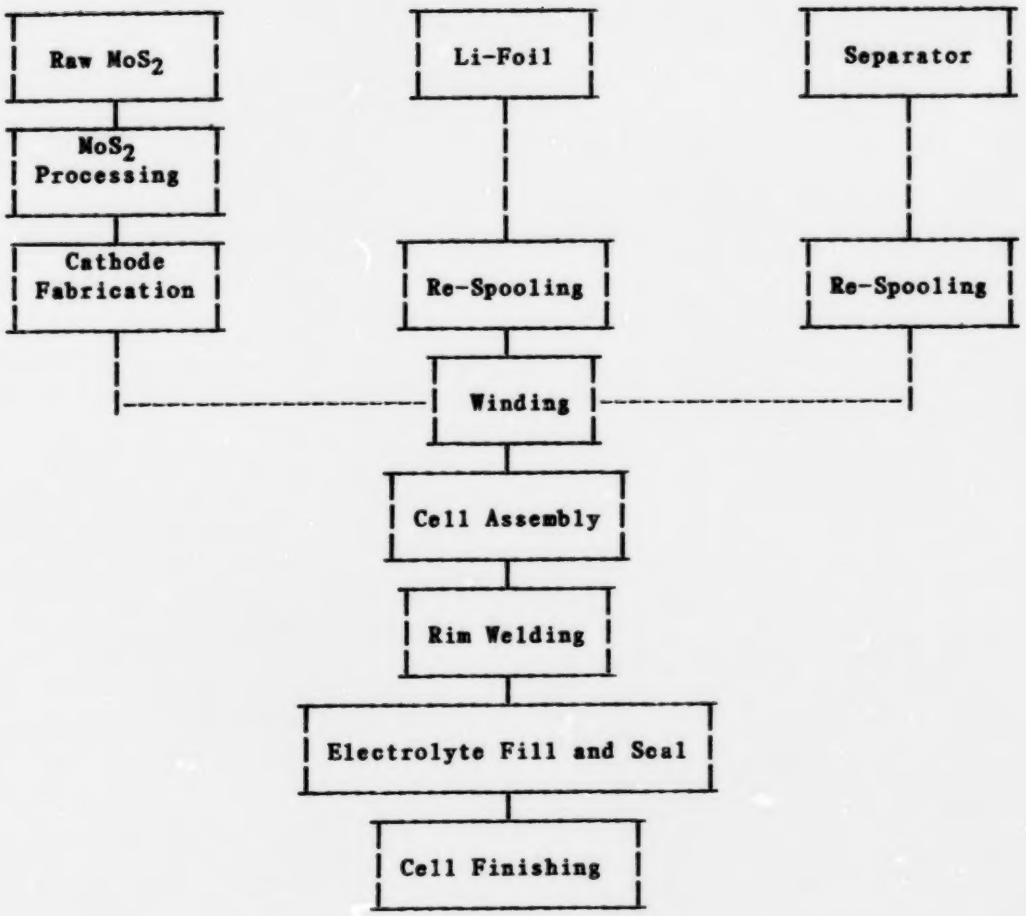

Figure 2: Block Diagram of 'BC' Cell Fabrication

3.2 CATHODE MACHINE

The main parts of the cathode machine and their functions were as follows.

A supply reel supplied a continuous strip of the current collector foil. A spreader applied the active material to one side of the foil in the form of a slurry containing MoS_2 in powder form. The wet cathode strip is dried in an oven and wound onto a take-up spool.

The existing cathode machine, originally designed for 'C' cell cathodes, was modified in order to be able to produce wider cathodes for 'BC' cells. These modifications include a new spreader, slurry tank, circulation pump, and slurry metering device.

3.3 LITHIUM SPOOLER

Lithium foil in battery grade purity is commercially available in dimensions suitable for the 'BC' size cells. However, the spools the foil is supplied on and the alignment of the foils on the spools is not suitable for winding of the jelly rolls. Therefore, a lithium spooler was built which respooled the foil from the supplier's reel to a precision reel to be used with the cell winder.

3.4 STORAGE CHAMBERS

Lithium batteries have to be assembled in a dry room atmosphere due to the sensitivity of active materials to water vapour. Assembly of the 'BC' cells was undertaken in Moli Energy's dry room which is kept at a relative humidity of 2% or lower. Past experience with smaller cell sizes has shown that even under these conditions the time of exposure of anode and cathode material to the dry room atmosphere has to be

limited. Therefore, two storage chambers were built to keep the active materials in an dry argon atmosphere or under vacuum.

3.5 WINDER

The 'BC' cell winder was designed based on concepts developed for a manual 'C' size cell winder. The active materials and the separators which form the web were unwound from a total of five spools onto a motor driven mandrel. Air pressure operated disc brakes on the spools were used to maintain a minimum tension on the cathodes and separators. During the wind, tabs and insulators were inserted manually and spot welded to the lithium anode and the cathode current collector.

Alignment of the components of the web was achieved by manually adjusting the positions of the spools with respect to the mandrel.

3.6 DIES AND DIE SETS

For the first 'BC' cell design, a deep drawn stainless steel can was available which could be used without modifications. Dies were only needed to form the cap blank which was to hold the two terminals, a fill tube, and a safety valve. The safety valve, which had been developed for this purpose, was a combination of a rupture disc and a spring loaded re-sealable vent. The vent pressure was 300 psi.

While the first 'BC' cell design was being tested, a new vent system had been developed for smaller cells. Vents are formed by stamping a 'C' shaped score into the bottom and top of the cold-rolled steel cell case. By controlling score diameter, score depth, and the shape of the score cross section, reproducible venting pressures were achieved. This development was transfered to the 'BC' size cells. New dies were necessary to form the vents in the cell case bottom and the cap.

3.7 GLASS-TO-METAL SEALS

In order to guarantee the hermeticity of the cell casing, a glass-to-metal seal was developed for the cathode feed-through. In order to be able to fuse the glass insulator and bond it to the outer ferrule and the inner post, a tube furnace was purchased. The furnace controlled the rate of heat-up and cool down as well as the working temperature at which the seal was formed.

3.8 CAN WELDER

Cell caps were welded to the can using T.I.G. welding. After several trials it was established that the best results could be achieved using a pulsed power supply. A Powerweld Engineering model PW 102 power supply was purchased for this purpose.

Other welds were needed to join the terminals and the fill tube to the cap. In all cases, the welds were circular and a rotary table was used. Special adapters were made for each part to hold it in place during welding.

3.9 ELECTROLYTE FILLING

Cells were filled with electrolyte after assembly and then hermetically sealed by closing off the fill tube. The 'BC' filler consisted mainly of a vacuum system and an electrolyte handling system which is designed to avoid contamination of the electrolyte with air.

For wetting, the cell was connected to the wetter and evacuated. The cell was then backfilled with electrolyte pressurized by argon. The cell was sealed either with a gas tight valve, or capped with a fitting.

3.10 BUNKERS

In any energy storage device with a high energy storage density and a capability of delivering the stored energy at high rates, some danger exists in the event of an uncontrolled release of energy. With a new product, special care has to be taken until the conditions of a safe assembly and operation are defined. Therefore, a bunker with four rooms was built. The first room housed the cell wetting operation, the second the performance testing, the third the safety testing, and the fourth cell disposal. The bunker was equipped with forced ventilation and designed to withstand the maximum pressures which could be expected from the simultaneous venting of 6 'BC' size cells in one of the rooms. The bunker roof was designed to lift off in order to provide an emergency pressure relief.

All battery testing can be controlled remotely from a separate control room. Safety experiments can be monitored using closed circuit T.V. equipment. The bunkers were equipped to monitor cell voltages, temperatures, and pressures.

3.11 PERFORMANCE TEST EQUIPMENT

The performance test equipment was designed to determine both cell and battery performance parameters. The equipment can handle batteries of up to 6 cells in series.

Several cycle testers were built to determine cell capacity and cycle life. Completed were one cycler with 50 A current capability, one cycler with 20 A current capability, and four cyclers with 10 A current capability. All units have independently setable charge and discharge currents over a range from 10 percent to 100 percent of fullscale. Switching from charge to discharge and vice-versa is automatically performed at setable battery voltages. The units are equipped with a fail-safe device that interrupts the current if the voltage on any of

the cells in a battery pack is outside a given range. The cell voltages are continuously monitored and can be recorded using a chart recorder. All cyclers can be interfaced with a mini computer for future work.

A Kikusui model PLZ 700 W programmable load was purchased for determining the peak power outputs of 'BC' batteries. It has a current capability of up to 140 A.

Several environmental chambers were bought and set up in order to perform the tests at a controlled temperature. The chambers are setable in a range from $-10^{o}C$ to $+50^{o}C$.

3.12 SAFETY TEST EQUIPMENT

The electrical safety testing comprised overcharge tests, forced discharge tests, and short circuit tests. For the former two tests, cycle testers were used with their fail-safes defeated so that the voltage on a single cell can be between the limits of +15 V and -15 V. For the short circuit test, a high current relay was used which was activated from the control room. The impedance of this circuit, including a shunt resistor which is used to determine the short circuit current, was about 3 mΩ.

The mechanical testing comprised shock and vibration. For the shock tests, a cell was mounted on a holder which could move along a short track. Acceleration of the holder down the track was achieved by a heavy pendulum swinging against shock absorbers mounted on the holder. Peak accelerations of up to 1500 ms^{-2} were achieved as measured by an accelerometer.

For the vibration tests, the cells were mounted onto a vibration table which was sinusoidially moved out of its rest position with a fixed

amplitude of .076 cm by a cam-shaft arrangement driven by a DC electric motor. The vibration frequency was controlled by controlling the motor speed. A controller allowed the frequency to vary at a setable rate in a range from 5 Hz to 55 Hz.

During all safety tests, cell voltage, cell current, cell temperature, and pressure were continuously monitored. A data logging system collected the data and stored them on a hard disk, which is part of a Zilog mini computer. Software developed for other projects was used for data evaluation on this computer.

3.13 CELL ASSEMBLY AND TESTING CAPABILITY

The above described assembly equipment allowed the assembly of about 2 'BC' size cells per day. Many of the assembly steps require a large amount of manual labour. However, the production rate was totally sufficient to supply cells for testing. A single cell cycle life test, assuming 100 life cycles, a charge time of 10 hours, and a discharge time of 3 hours required one cycle life tester for 54 days. With 6 testers available now, the test capacity was about 1 cell or battery every 9 days.

4 CELL AND BATTERY FABRICATION AND TESTING

4.1 CELL FABRICATION

A total of 18 'BC' cells were fabricated during the period of the contract. This number does not include cells that were rejected at various inspection points, but only includes cells that were filled with electrolyte. Most failures before filling were assembly failures caused by internal shorts.

Table 2 gives a summary of the fabricated cells and their use in the test program. Of these cells, 2 failed due to a high internal impedance after wetting with electrolyte. It is speculated that the high impedance was due to incomplete filling of the cells with electrolyte. One cell shorted internally during the first recharge. The reason for this failure is not known but it is most likely an assembly error.

Table 2
Summary of Cells Fabricated and Their Use

CELL ID	CELL TYPE	PERFORMANCE TESTS	SAFETY TESTS	COMMENTS
840425-1	1	-	Short circuit	-
840425-2	1	Cycle test to 50 cycles	-	-
840622-1	1	Cycle test to 68 cycles	-	-
840717-1	1	Battery testing	Shock + vibration	2 cell battery
840719-1	1	Cycle test to 15 cycles	Short circuit	-
840824-1	2	Cycle test to 22 cycles	Forced discharge Overcharge	-
840824-2	2	Cycle test to 107 cycles	-	-
840927-1	2	Battery testing	-	2 cell battery
841005-1	2	-	-	Cell showed high internal resistance
841016-1	2	-	-	-
841018-1	2	-	-	Cell showed high internal resistance
841025-1	2	-	-	Cell shorted internally on first recharge
841018-2	2	Battery testing	-	6 cell battery
841022-1	2	Battery testing	-	6 cell battery
841023-1	2	Battery testing	-	6 cell battery
841023-2	2	Battery testing	-	6 cell battery
841024-1	2	Battery testing	-	6 cell battery
841025-2	2	Battery testing	-	6 cell battery

4.2 BATTERY FABRICATION

Two batteries were built using the 'BC' cells, one with 2 cells, the other with 6 cells in series. The cells were held in place and electrically insulated from each other by a plexiglass base and top.

Terminals were provided so that the individual cell voltages could be measured.

Table 3 gives the physical properties of the 6 cell battery.

Table 3
6 Cell 'BC' Battery

Battery Dimension	:	25 cm x 16 cm x 21 cm
Battery Weight	:	7.3 kg
Nominal Battery Voltage	:	10.8 V
Nominal Battery Capacity	:	47.6 Ah

4.3 SINGLE CELL PERFORMANCE TESTING

4.3.1 Results of Cycle Life Tests

All cycle life testing was performed by recharging the cells at constant current to a set charge voltage cutoff, then immediately discharging the cells at a constant current to a set discharge voltage cutoff. For the charging, a current of 5 A was chosen. This corresponds to a current density of about .3 mA cm^{-2} at the 'BC' cell electrodes. Experiments with small cells have shown that charge current densities higher than .35 mA cm^{-2} can lead to cell performance degradation.

For the purpose of these experiments, the discharge current was set at 15 A. This corresponds to a .32 $hour^{-1}$ rate, i.e. a discharge of the nominal capacity of 47.6 A hours in 3.2 hours. This particular rate was chosen as it represents the average rate of discharge of a battery in a typical electric vehicle application. The environmental temperature for these tests was 20°C.

Figure 3 shows the cell voltage of a 'BC' size cell as a function of time for a complete charge/discharge cycle. The voltage variation of the cell with the state of charge on charge and discharge is due to a change in the chemical potential of the Li_xMoS_2 intercalation compound. This feature can be used as a state of charge indicator.

The dependence of the cell voltage on the state of charge also allows the control of the depth of discharge by adjustment of the cutoff voltages for charge and discharge.

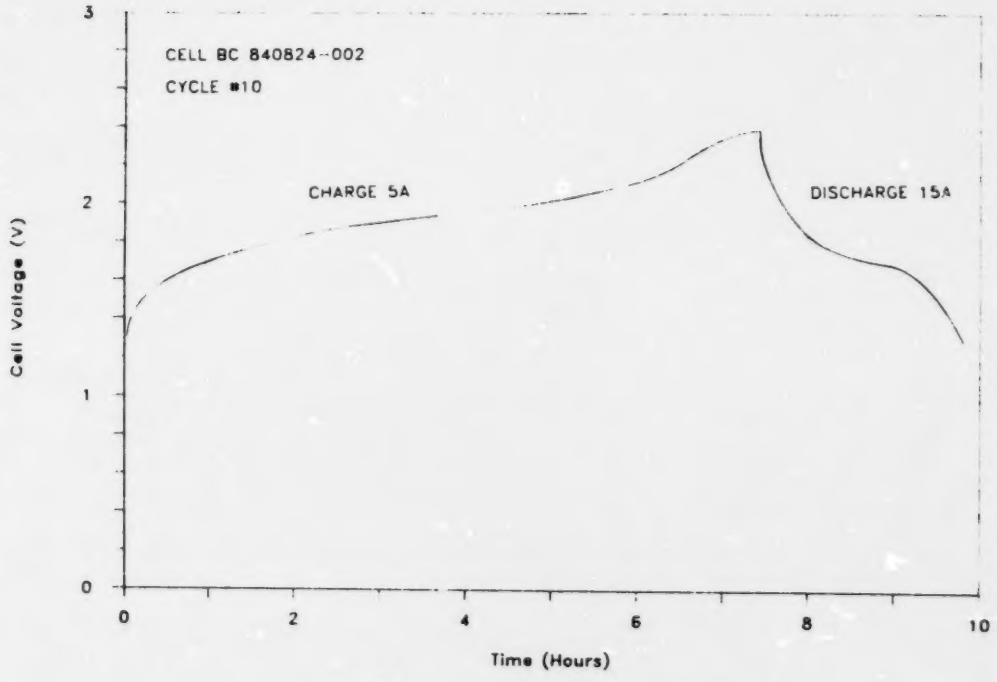

Figure 3: Charge and Discharge Voltage Voltage Profile of a 'BC' Cell. Charge 5 A, Cutoff 2.4 V. Discharge 15 A, Cutoff 1.3 V.

Figure 4 is a plot of the achieved capacity as a function of cycle number for a type I cell. These types of plots will be called cycle life plots in the following. The cutoff voltages were set at 2.4 V and 1.3 V. Within these limits a capacity of 41.5 Ah was realized on the first cycle. There is some fade of capacity, with 64% of the first cycle capacity remaining on the 50th cycle. The test was terminated after cycle 50.

Figure 5 contains the cycle life plot for a type II cell cycled under the same conditions. The two cell types have the same first cycle capacity, however, the type II cell shows a lower capacity fade with 78% of the initial capacity remaining on cycle 50. On cycle 55, the voltage cutoff on discharge was lowered to 1.1 V for a single cycle. The capacity on that cycle was 44.5 Ah.

Figure 5 also shows the effect of a change in the cutoff voltages on the achievable capacity. This type II cell was charged to 2.6 V and discharged to 1.1 V. A capacity of 66.5 Ah was achieved on cycle 1. Continued cycling under these conditions resulted in a capacity of 50.5 Ah on cycle 22. The average voltage on discharge under these conditions is 1.7 V, thus the total energy delivered on the first discharge is 113 Wh. The test was terminated after 22 cycles. The cell was then used for safety tests.

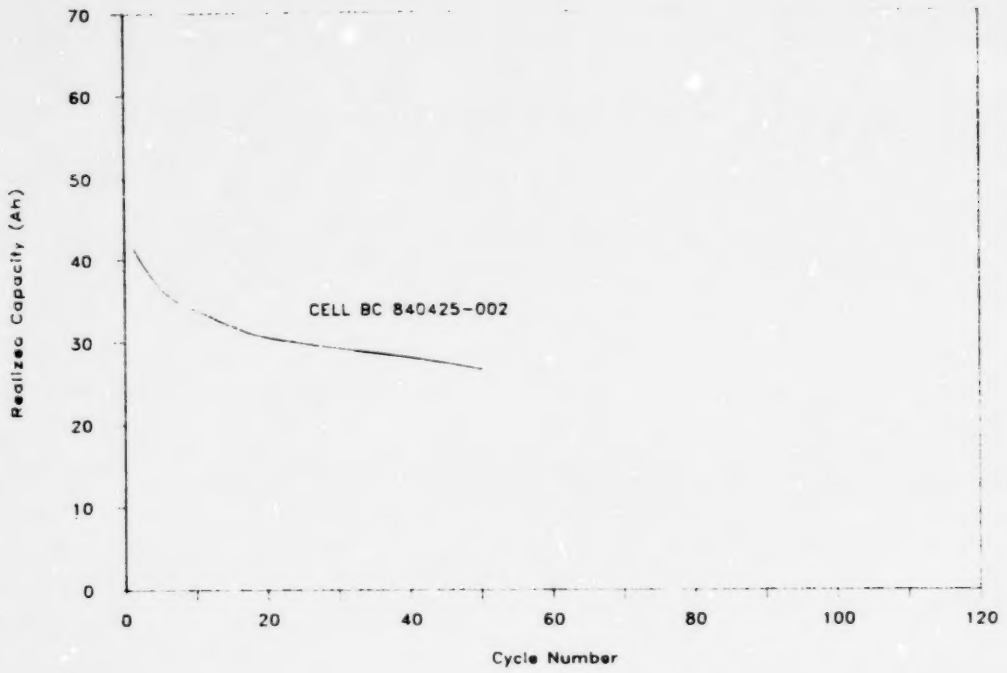

Figure 4: Cycle Life Plot for a Type I Cell. Charge 5 A, Cutoff 2.4 V. Discharge 15 A, Cutoff 1.3 V.

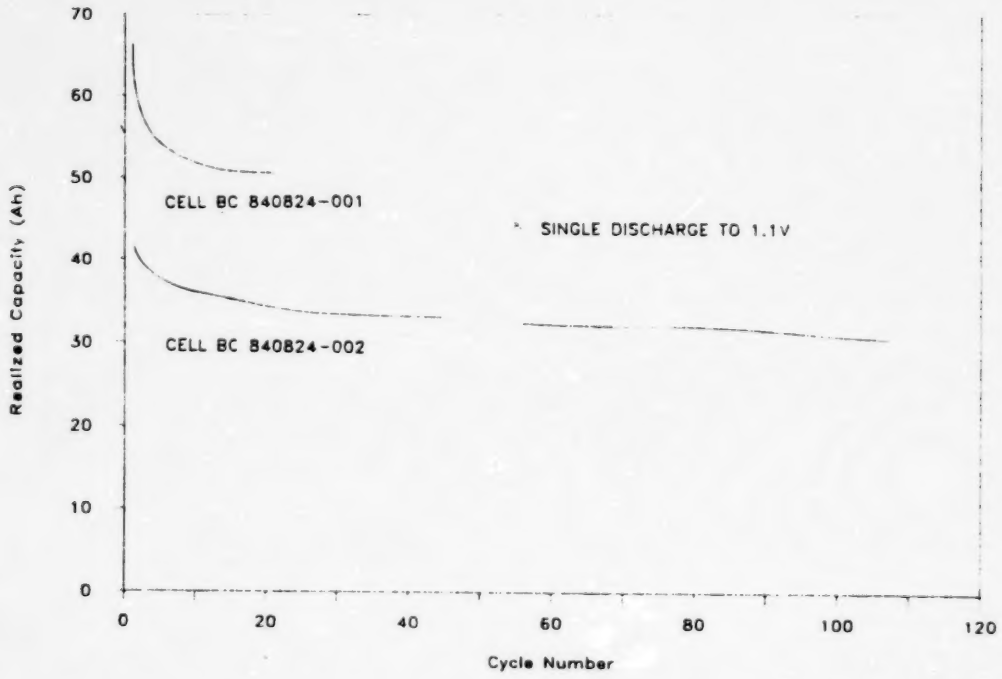

Figure 5: Cycle Life Plots for two Type II Cells. Cell 'BC' 840824-001: Charge 5 A, Cutoff 2.6 V. Discharge 15 A, Cutoff 1.1 V. Cell 'BC' 840824-002: Charge 5 A, Cutoff 2.4 V. Discharge 15 A, Cutoff 1.3 V.

4.3.2 Determination of Cell Impedance

The rate capability of the 'BC' cells is determined in part by their internal impedance. The impedance of a 'BC' cell was measured using frequency response analysis. With this technique, a small sinusoidal alternating current signal is applied to the cell. The response of the cell voltage is determined and the complex impedance of the cell for the frequency of the exiting alternating current signal is calculated from the ratio of the voltage and current signal amplitudes and their phase shifts. With the particular instrumentation used at Moli which is based on a Solartron Model 1172 frequency response analyzer, the frequency can be swept from 10 kHz to .1 mHz in setable steps. The results are usually represented in a Cole-Cole plot, as in Figure 6.

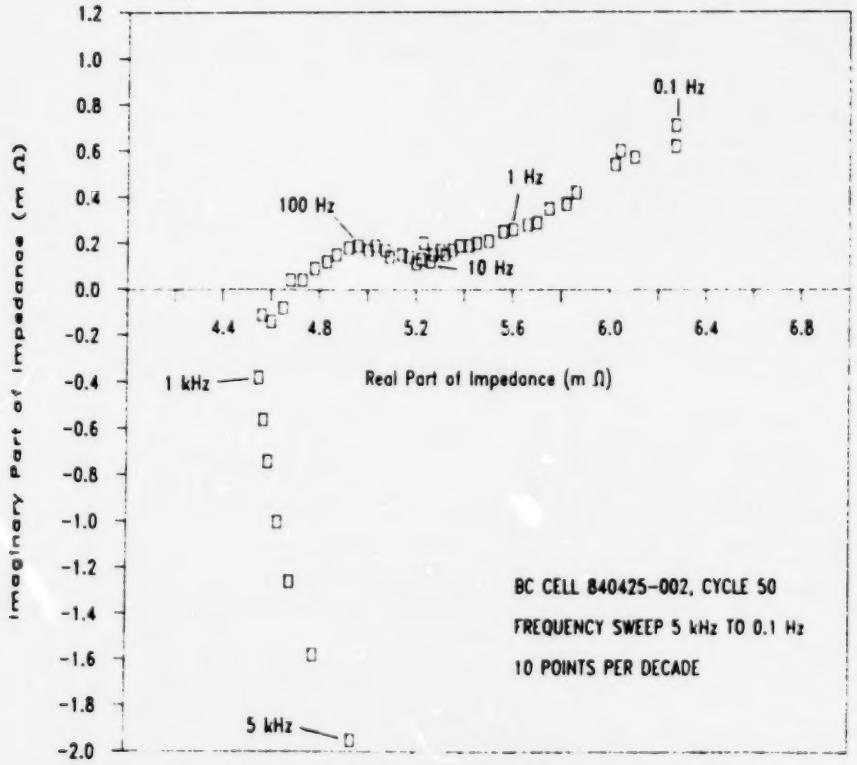

Figure 6: Cole-Cole Plot of Impedance of a 'BC' Cell

Figure 6 gives the complex impedance for 'BC' cell # 840425-002 at cycle # 50. The cell is at a voltage of 1.62 V, i.e. it is about 1/4 charged. The interpretation of the spectra presented here is based on studies published in the literature (2) as well as on previous studies of the frequency response of anode and cathode half-cells (3).

Three regions can be easily identified in the cell's frequency response. The first region which extends from frequencies of about 1 kHz to higher frequencies is dominated by the inductance of the cell winding. This inductive part will only affect the performance of the cell for pulsed loads of very short (< 1 ms) duration. The cell impedance at the intercept with the real axis, R_{ohm}, is about 4.5 mΩ. Contributions to this ohmic impedance arise from the resistance of feed-throughs, tabs, current collectors, and the electrolyte.

The next region, which extends in frequency down to about 10 Hz, is characterized by its near semicircular shape in the Cole-Cole plot. The diameter of this semicircle, Ri, is about .7 mΩ. This shape arises from resistive and capacitive elements in series. The contributions to the impedance in this region arise from the charge transfer at the interfaces of the electrodes with the electrolyte, from the ionic transport in the pores of the cathode, and from the resistance between cathode active material and the current collector.

The third region which extends into the low frequency region is called the mass transport region. It is dominated by the lithium ion diffusion in the electrolyte and in the solid Li_xMoS_2 intercalation compound.

4.3.3 Discussion of Single Cell Performance Testing Results

The performance of the 'BC' cell can be evaluated by comparison with 'C' cell performance. Table 3 gives this comparison using a scaling factor of 22 for the ratio of the active materials of the two cell sizes.

Under standard cycling conditions the 'BC' cells fall about 13% short of the expected capacity values. This short fall might in part be due to the somewhat higher than expected values for the cell impedance. Increased cell impedance causes shallower discharges since the cell voltage under load is used to determine the endpoints of charge and discharge. The increase in cell impedance is probably due to the impedance of the cell terminals and connections between terminals and the anode and cathode current collectors which do not scale between the two cell sizes.

Table 3
Comparison of 'BC' Cell Performance with 'C' Cell Performance

Note: Expectation values for the 'BC' cells were calculated using a ratio of active material of 22:1 for 'BC' and 'C' cells.

	'C' CELL	'BC' CELL	
		Expected	Actual
Capacity, 1st cycle, standard conditions	2.16 Ah	47.6 Ah	41.5 Ah
Capacity, 100th cycle, standard conditions	1.60 Ah	35.2 Ah	30.5 Ah
Capacity, 1st cycle, wide voltage limits	3.00 Ah	66.0 Ah	66.5 Ah
Cell impedance, 1 kHz, cycle 50	60 mΩ	2.7 mΩ	4.5 mΩ
Energy density, standard conditions	60 Wh/kg	78 Wh/kg	68 Wh/kg
Energy density, wide voltage limits	78 Wh/kg	102 Wh/kg	103 Wh/kg

Over the wider voltage limit, an energy density in excess of 100 Wh/kg has been achieved. With further engineering efforts, a reduction of the cell weight of 100 g to 200 g should be possible which would result in a 10 to 20% increase in the gravimetric energy density.

4.4 SINGLE CELL SAFETY TESTING

4.4.1 Short Circuit Tests

Short circuit tests were performed on two cells by connecting the cell terminals to a load of about 3 mΩ impedance. During the tests, the cell current, the gas pressure inside the cell, and the temperature of the cell case at two points, one point close to the top, the other close to the bottom, were monitored.

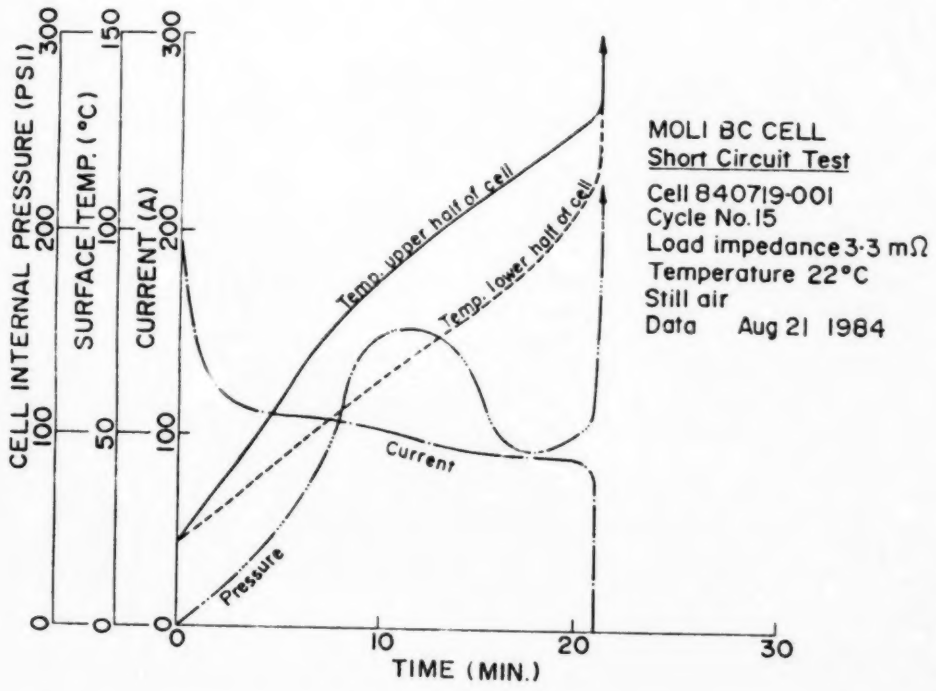

Figure 7: Cell Gas Pressure, Surface Temperature, and Short Circuit Current for a 'BC' Cell Shorted through a 3.3 mΩ Load at Time 0.

Figure 7 shows the results of a test performed on cell #840719-001 after 15 standard cycles. The cell had an initial short circuit current of about 200 A. The current then dropped to an average of about 100 A which was maintained for more than 20 minutes. During that time, the case temperature rose continuously at a rate of about 5°C per minute. The internal gas pressure rose first to about 160 psi and then dropped to about 100 psi. Analysis of the video movie made of this event showed that this drop was due to pressure release through a cracked glass-to-metal feedthrough. About 21 minutes after initiation of the short, the current fell suddenly to zero, indicating an internal short. Within several seconds, the cell vented by opening both the top and bottom vent. The vent was associated with flames due to burning gas and some ejection of solid material from the jelly roll. The case was left intact other than the opened safety vent.

The other short circuit test performed on cell #840425-1 showed similar results.

4.4.2 Overcharge and Forced Discharge Tests

Both overcharge and forced discharge tests were both conducted on cell #840824-1 after 20 standard cycles. The tests were performed at an ambient temperature which varied between 17°C and 11°C. Figure 8 shows the results of the test. The cell was charged at 5 A for 4.5 hours past the charge endpoint. For the first 2 hours, the voltage rose slowly to 2.7 V. During that time, the cell case temperature did not rise and the gas pressure remained at atmospheric pressure. After 10 Ah of overcharge, voltage, temperature, and pressure rose sharply. The test was terminated at 22.5 Ah of overcharge.

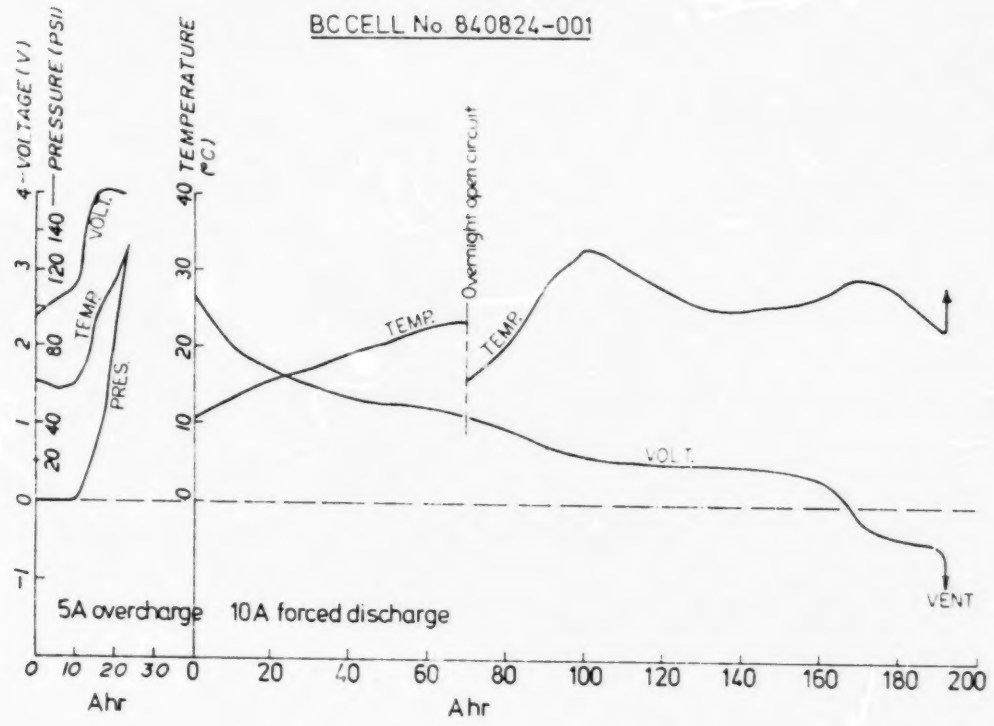

Figure 8: Cell Gas Pressure, Case Temperature, and Voltage for a 'BC' cell First Overcharged at 5 A, then Discharged at 10 A.

The forced discharge test was performed following the overcharge test. The cell appeared not to have been damaged severely by the previous abuse as it achieved 44 Ah of capacity to 1.3 V. The discharge was continued beyond this point. Voltage reversal did not occur until 168 Ah of charge was passed. Before voltage reversal, the cell case temperature did not rise to more than about 20°C above ambient. After voltage reversal, the cell stabilized at first at a voltage of -.6 V, then voltage and temperature rose sharply. The vent occured about 2.4 hours into voltage reversal. The severity of the vent was comparable to the vent observed at the end of the short circuit test. The percentage of forced discharge achieved before venting was approximately 300% of the nominal capacity.

4.4.3 Shock and Vibration Test

Shock and vibration tests were performed on cell #840717-001. During the shock test, a cell was subjected to a peak acceleration of 1500 ms^{-2} and an average of 800 ms^{-2} over a time period of 3 ms. The test was performed by accelerating both parallel and perpendicular to the cylinder axis of the cell. No damage occurred to the cell.

The vibration test was carried out over a frequency range from 5 to 55 Hz. Starting from 5 Hz, the frequency of the vibrations was increased at a rate of 1 Hz/minute to 55 Hz and then decreased to the lower limit. The amplitude of the vibrations was .76 mm. The test was performed with the vibrational amplitude both parallel and perpendicular to the cylinder axis of the cell. No damage occurred to the cell. After being subjected to these tests the cell was used in the two cell battery pack.

4.4.4 Interpretation of Safety Testing Results

The safety test results are best interpreted by comparison with 'C' cell data, some of which have been published elsewhere (4).

In (4) it has been shown that the outcome of short circuit tests and forced discharge tests of 'C' cells depends very much on thermal management, that is on the capability to remove heat from the jelly roll so that the internal cell temperature does not exceed a critical value. It is therefore not surprising, that safety problems are more likely to occur in larger cells with longer paths for heat conduction and a smaller surface area to volume ratio.

The cell used for the short circuit test had an open circuit voltage of 2.2 V. Shorted through a 3.3 mΩ load, the peak current was 200 A.

This corresponds to an effective cell impedance of 7.7 mΩ which is higher than the impedance determined by frequency response analysis of about 5.5 mΩ for a frequency of 1 Hz. The reasons for this discrepancy are not known. Experiments with 'C' cells have shown that the cell impedance at 1 Hz can be used to calculate the peak current under short circuit conditions. For example, a typical 'C' cell on its 10th cycle has an impedance of 80 mΩ at 1 Hz and a short circuit current of about 27 A.

Using the 'BC' cell data, the rate of internal heat generation was calculated to be about 320 W initial, dropping to about 150 W after several minutes. In case of a thermally insulated cell, (adiabatic case), this would cause a rise of cell temperature of 8°C per minute, assuming a value of 1 joule/°C kg for the specific heat of the cell. This value of the specific heat was determined experimentally for 'C' cells. With an environmental temperature of 10°C, the cell should have reached 180°C, the melting point of lithium, in 21.25 minutes. This time corresponds well with the time it took for the shorted cell to vent in the experiment conducted. The molten lithium caused the cell to short internally which leads to venting due to rapid decomposition of the electrolyte.

In small cells, like 'AA' size cells, the melting point of the lithium is never reached due to a sufficient rate of heat transfer from the active material to the case and to the environment. In 'C' size cells the result of this short circuit test depends critically on the heat transfer from the case to the environment. Cells with insufficient heat transfer vent similar to the 'BC' cells.

Compared to a 'C' cell, the ratio of surface area to volume for a 'BC' cell is less by a factor of 2.8. Furthermore, the path length for heat conduction from the center of the cell to the case is longer by a factor of 2.5. Therefore, the core of a 'BC' cell can probably be treated as an adiabatic case, which justifies the analysis of the short circuit results presented above.

The result of the overcharge and forced discharge tests were similar to 'C' cell results. 'C' cells have shown no ventings on overcharge at less than 90% overcharge beyond the fully charged state.

On forced discharge beyond the normal end of discharge point, the excess lithium incorporated into the cell design continues to intercalate into the molybdenum disulfide. Thus typically 300% overdischarge can be achieved before cell reversal occurs. Venting occurs if the overvoltage on reversal is sufficiently high.

In summary, the behaviour of 'BC' cells under electrical and mechanical abuse conditions agrees with extrapolations made from 'C' cell behaviour. In the short term, improvements in safety behaviour will most likely come from external devices like fuses and diodes. Several solutions to the thermal management problems of large Molicels^{TM} under abusive conditions are being discussed at present and will be evaluated in the near future.

4.5 BATTERY PERFORMANCE

4.5.1 Cycle Testing and Stored Energy

The two batteries tested consisted of one pack of 2 cells in series and another of 6 cells in series. Both batteries were cycled under conditions similar to that of the single cell cycling. The voltage of the battery was used to determine the end points for charge and discharge. After the initial assembly of the batteries, no effort was made to equalize the state of charge between individual cells.

The 2 cell battery was charged at 5 A to 4.8 V and discharged at 15 A to 2.6 V. A capacity of 38 Ah was achieved on the first cycle and the capacity on the 11th cycle was 32 Ah. The test was terminated after cycle 11.

The 6 cell battery was, for most of its cycle life test, charged at 5 A to 14.4 V and discharged at 15 A to 7.8 V. A capacity of 42 Ah was achieved on the first cycle. This corresponds to an energy storage capacity of 453 Wh. Figure 9 shows the voltage profile for cycle 20 when a capacity of 35.5 Ah was achieved. For cycles 25 to 28, the voltage limits were changed to 15.6 V and 6.6 V, respectively. Figure 10 shows the voltage profile for cycle 26 when a capacity of 56 Ah was achieved. This corresponds to an energy storage capacity of 570 Wh.

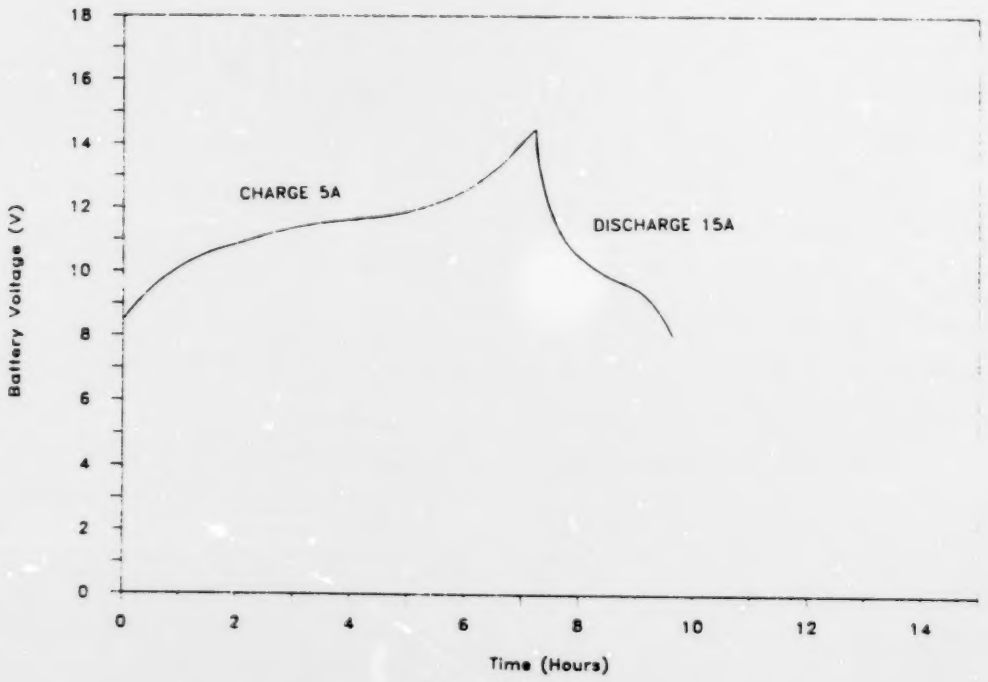

Figure 9: Voltage Profile of the 6 Cell 'BC' Battery. Charge Cutoff Voltage 14.4 v, Discharge Cutoff Voltage 7.8 V.

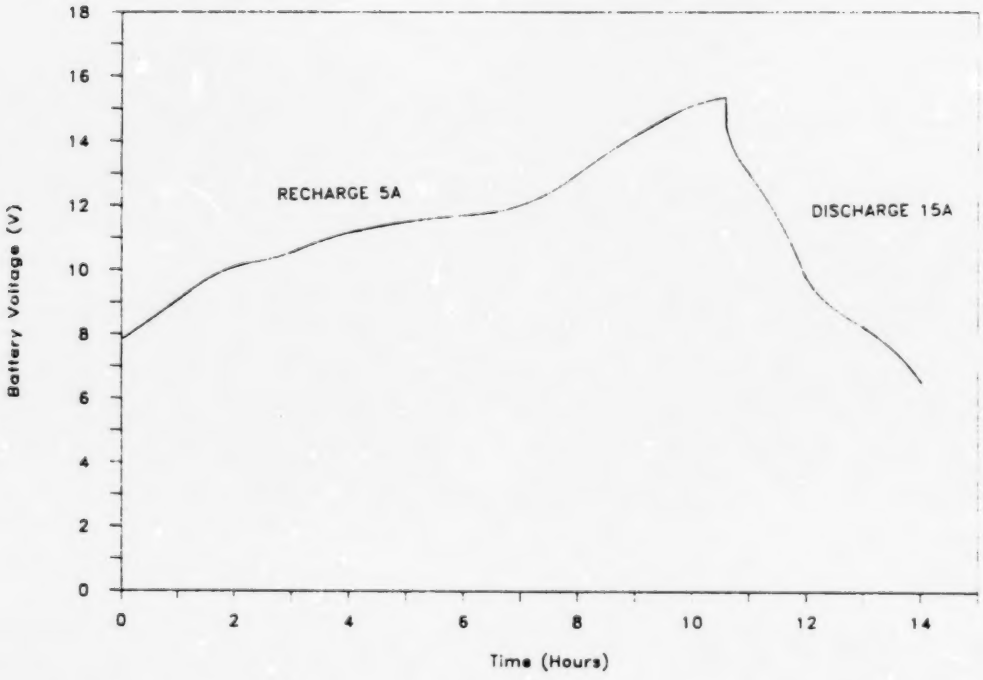

Figure 10: Voltage Profile of the 6 Cell 'BC' Battery. Charge Cutoff Voltage 15.6 V, Discharge Cutoff Voltage 6.6 V.

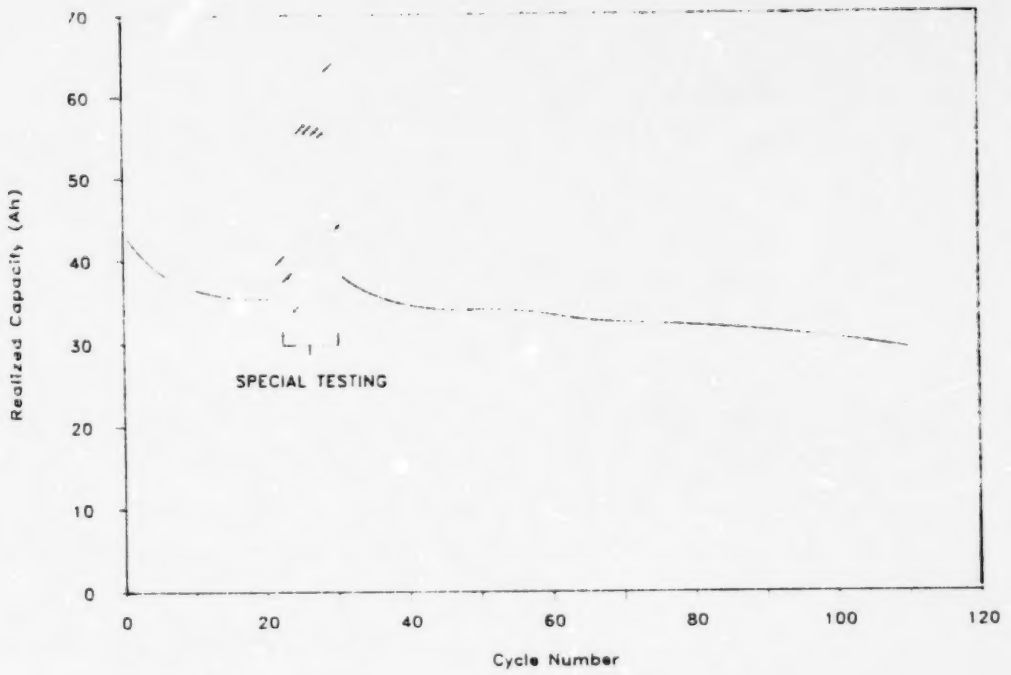

Figure 11: Cycle Life Plot for the 6 Cell 'BC' Battery Standard Testing conditions were: Charge 5 A, Cutoff 14.4 V, Discharge 15 A, Cutoff 7.8 V.

Figure 11 shows the realized capacity for this battery as a function of cycle life. At cycle 100, the capacity is 30.5 Ah, which is 73% of the initial capacity. The test was terminated at cycle 110. The battery is still operational and can be used for demonstration purposes.

During cycling, the voltage of individual cells was monitored. Some imbalance in the state of charge of individual cells developed during the course of the experiment, which caused the cell voltages to slowly drift apart. Table 4 gives the individual cell voltages after 100 cycles in a fully charged state of the pack.

Table 4
Voltage of Individual Cells of the
6 Cell Battery Pack After 110 Cycles

(Battery is in the charged state)

Position of Cell	Cell Voltage
1	2.135
2	2.042
3	1.999
4	2.481
5	2.125
6	2.216

4.5.2 Continuous Power Capability

The continuous power capability of the 6 cell battery was assessed by discharging the fully charged battery to a fixed voltage limit at various currents. The power delivered thus varied with the battery voltage and was smallest at the end of the discharge. A constant power discharge was not possible with the equipment available at the time. A standard continuous power capability test was conducted as follows. The battery was charged at 5 A until the battery voltage reached the upper cutoff voltage. The battery was then discharged at the highest current of the experiment, which in this case was 50 A. The discharge was terminated when the battery voltage reached the lower cutoff voltage. The battery was then rested on open circuit for a time equal to that required for the just completed discharge. The battery was then further discharged at a discharge current equal to one half of the current on the previous discharge again until the cell voltage fell to the lower cutoff voltage. The delivered battery capacity at this lower rate was then taken to be the sum of the discharge capacities for the two discharges. The process was repeated until the discharge rate had been reduced successively by one half until the realized capacity at the lowest rate desired had been measured. This procedure was found to give equivalent results to those obtained when the battery was fully charged before each discharge at the desired rate. A plot of the realized capacity versus the discharge current is called a signature curve.

Figure 12 gives signature curves for the 6 cell battery for three different temperatures for a charge cutoff voltage of 14.4 V and a discharge cutoff voltage of 7.8 V. Prior to this experiment, the pack had been cycled 21 times. The change in realized capacity with temperature is small between 20°C and 0°C, it is however significant at -10°C for currents above 20 A.

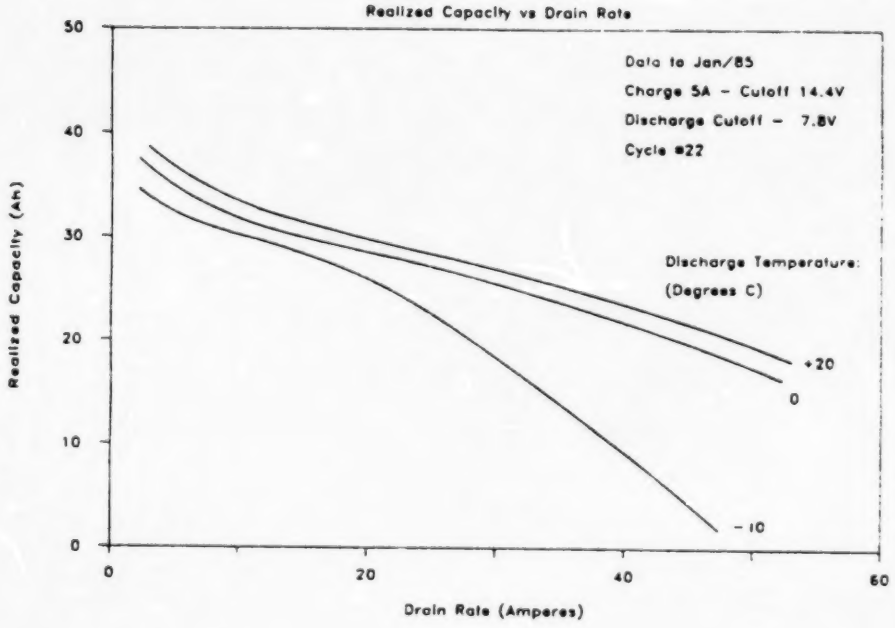

Figure 12: Signature Curves for the 6 Cell 'BC' Battery. Standard Voltage Limits.

Figure 13 gives a signature curve for wider voltage limits, i.e. a charge cutoff voltage of 15.6 V and a discharge cutoff voltage of 6.6 V. Widening the voltage limits resulted in a significant increase of the realized capacity. It increased by 60% to 63.5 Ah at a 2 A discharge current and by 58% to 32.5 Ah at a 50 A discharge current.

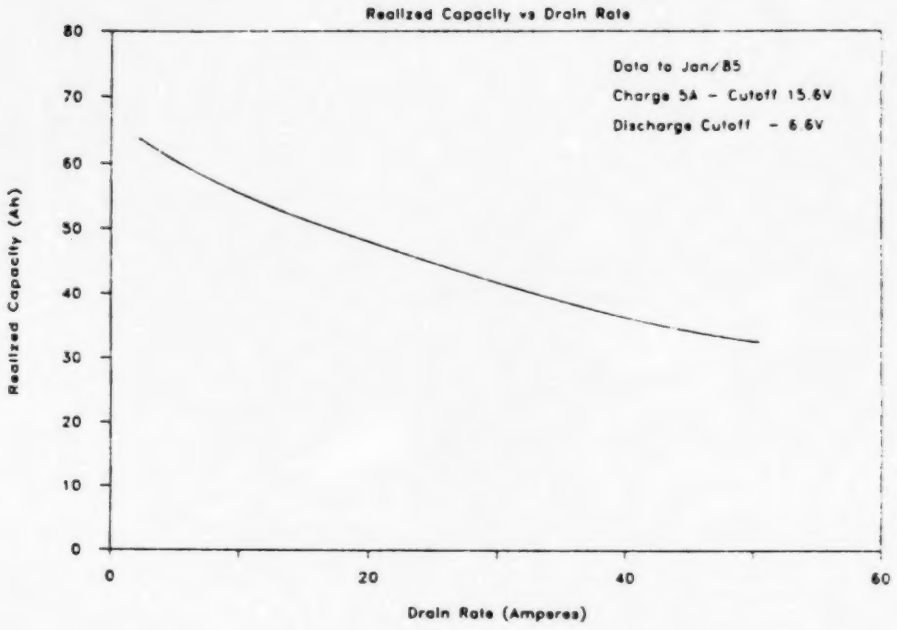

Figure 13: Signature Curve for the 6 Cell 'BC' Battery. Wide Voltage Limits, Temperature 20°C.

4.5.3 Peak Power Capability

For the purpose of this study, the peak power of the battery pack was defined as the maximum power that could be sustained for a duration of 5s for a fully charged battery.

The battery was charged to 15.6 V, then discharged through a variable load at different currents for 5s duration at each current value. After each discharge, the battery was on open circuit for about 1 minute. The battery power output was derived from the battery voltage under load measured at the end of each 10s discharge and the discharge current. Figure 14 shows the battery power output as a function of the discharge current. The peak power was 932 W. With a total battery weight of 7.3 kg this corresponds to a peak power density of 128 W/kg for a fully charged battery after 29 deep cycles.

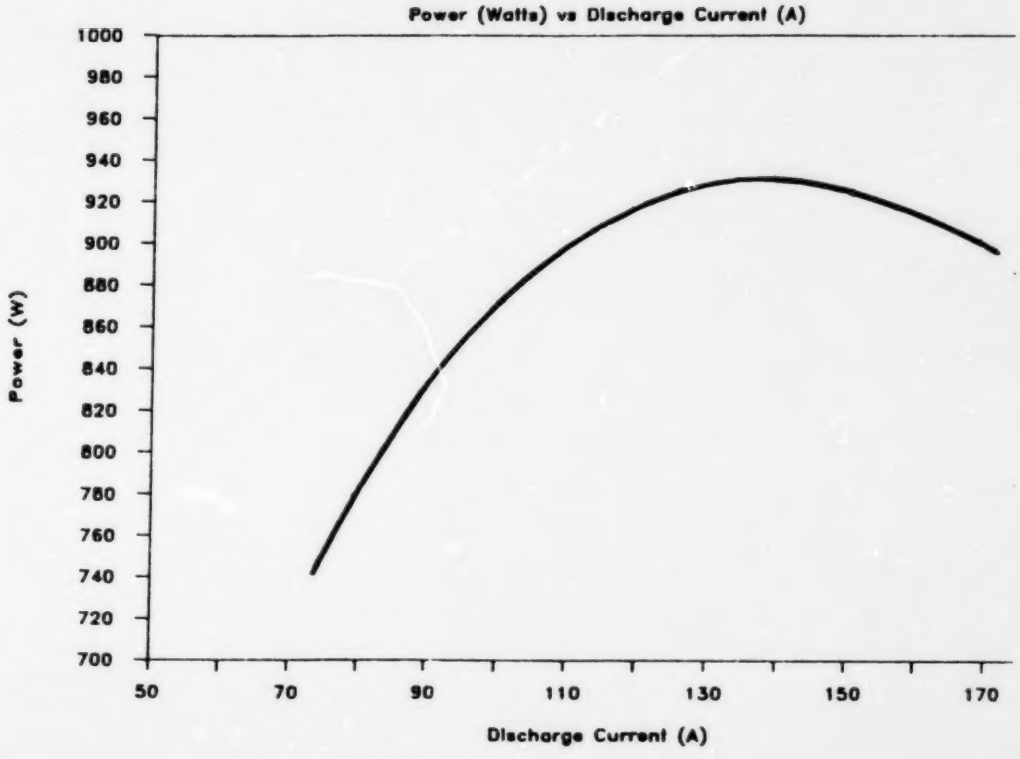

Figure 14: Power Output of a 6 cell 'BC' Battery at Various Current Drains.

4.5.4 Interpretation of Battery Performance Test Results

The battery performance test results are best interpreted by comparing them to the test results for single 'BC' cells and to 'C' cell batteries.

Comparison of the battery cycle test results with the single cell test results show that battery capacity and cycle life are similar to that of a single cell. Differences in individual cell capacities do not seem to effect battery cycle test results at least out to 110 deep cycles. The differences in the state of charge between cells after 110 cycles are probably due to various degrees of self discharge and differences in cell impedance and capacity fade from cell to cell caused by the lack of reproducibility in hand-assembled cells.

In Figure 15, energy density and power densities for the 6 cell battery are compared with single cell results and 'C' cell battery test results. The relative performance drop of the 'BC' battery compared to the single cell results is due to the increase of the battery weight over the weight of 6 individual cells caused by the additional battery hardware. Additional power losses are caused by the resistance of the inter-cell connectors. A comparison of the 'BC' battery energy and power densities with that of a 'C' cell battery shows the effects of scaling. In the scaleup from 'C' to 'BC' cells, the fraction of the weight of non-active materials to active materials has been reduced, resulting in an increase of energy density at low rates. At higher rates, however, the relatively larger resistive losses in current carrying elements of the large battery outweigh the weight advantages and cause a significant reduction in the power capability. From this analysis, it appears to be better to use a large number of 'C' cells in series over a corresponding smaller number of 'BC' cells if the particular application calls for high power density. However, from the point of view of reliability and cost, this is not a desirable alternative for large batteries.

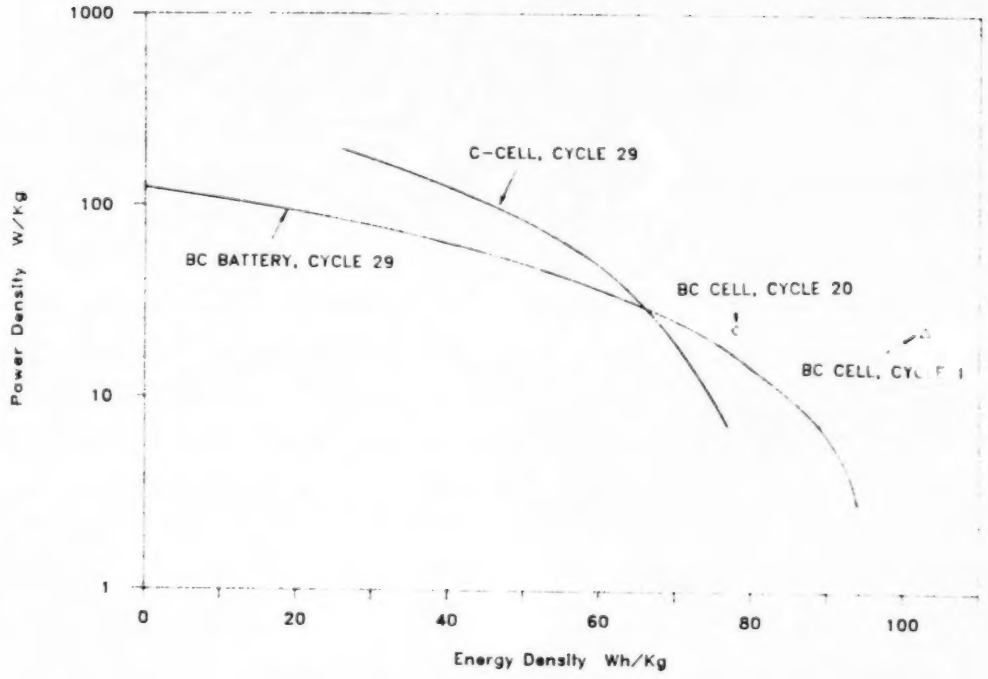

Figure 15: Relationship Between Gravimetric Power Density and Energy Density for a 6 Cell 'BC' Battery, Single 'BC' Cells, and 'C' Cells

This analysis points the way to an improvement in battery performance without changes to the chemistry of the system. Some weight saving could be achieved by the use of lightweight components in cell construction. The power density could be increased by optimizing the design of the current carrying elements of the battery. With these changes, an energy density in excess of 100 Wh/kg and a power density in excess of 150 W/kg should be achievable.

5 CONCLUSION

The scaleup of the lithium molybdenum disulfide battery technology from 'C' cell size to 'BC' cell size was performed successfully. The performance of the 6 cell battery with a maximum energy density of 94 Wh kg^{-1} and a peak power density of 128 W kg^{-1} nearly satisfies the requirements of an advanced electric vehicle design which is generally assumed to demand an average density of 100 Wh kg^{-1} and a power density of 150 W kg from the batteries. In comparison, advanced lead acid batteries developed for EV applications by programs supported of the D.O.E. of the U.S.A. achieved an energy density of 41 Wh/kg and a peak power density of 104 W/kg in 1984(5).

The analysis of the testing results shows that some further improvements to battery performance are possible by improvements to the cell design, especially in the area of power density. It is, however, clear that the power density of smaller cells does not scale well and that some penalty in this area is associated with the scaleup of the present spirally wound cell technology. A bipolar cell design would overcome these problems associated with the collection of large currents and should lead to the development of a battery system with power densities similar to small cells, i.e. of about 300 W kg^{-1}.

The performance of the system begins to degrade severely at temperatures below -10°C. In order to maintain a reasonable power density, the battery with the present chemistry has to be operated above this temperature. A significant improvement in this area will only be possible with the development of a new electrolyte system. In the case of an electric vehicle application at low ambient temperatures, the internal losses of the battery due to an increased impedance could be used to maintain an optimum operational temperate with proper thermal management of the battery pack.

The safety of the 'BC' cell has not been extensively evaluated in this project. More safety testing is necessary to define the envelope of safe operation for single cells as well as for batteries. The experimental evidence presented in this report suggests that the thermal management under abusive conditions is the key to controlling the effect of a battery abuse. Increasing the rate of heat conduction from the cell active material to the environment should reduce the tendency of 'BC' cells to vent under short circuit and other abusive conditions. A positive result is that, when cell venting was observed, the safety vents operated properly and the cell case was left intact. It appears to be possible to be able to contain a venting in a proper battery housing and thus minimize the effect of the vent on the battery environment.